

PATENT APPLICATION
IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of

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Naotaka IZUMI, et al. .

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For: METHOD FOR PRODUCING POLYURETHANE EMULSION FOR AQUEOUS ONE-
COMPONENT COATING AGENT

DECLARATION UNDER 37 C.F.R. § 1.132

Mail Stop Amendment
Commissioner for Patents
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Sir:

I, Takaki Jono, hereby declare and state:

THAT I am a citizen of Japan;

THAT I have received a M.S. degree in 1993 from Kumamoto University, Department of
Applied Chemistry;

THAT after receiving a M.S. degree from Kumamoto University, I joined Nippon
Polyurethane Industry Co., Ltd., where I worked on rigid and flexible foams and on coatings at
the Central Research Laboratory;

THAT I am a Technical Manager and the Waterborne Resin Research and Development
Group Leader at NPU Central Research Laboratory; and

THAT the following experiments were carried out by me or under my direct supervision.

EXPERIMENTATION

Production of Polyurethane Emulsion

An embodiment of the present invention (Example 4') was carried out on the basis of Example 4 of the present specification.

In Example 4', a polyurethane emulsion was produced in accordance with the disclosure in paragraphs [0058] and [0061] of the specification as filed. Although there is a difference in solid content, viscosity and average particle diameter from those of Example 4 as shown in Table 1 on page 30, paragraph [0065] of the specification, the differences are within the level of error. Thus, Applicants submit that the reproduced PU-4 is substantially the same as PU-4 disclosed in the specification as filed.

In Comparative Examples, PU-10 and PU-11 were produced by following the procedures of Example 4, except for replacing NCO-2 used as the *nonionic* isocyanate with the below-described NCO-3A and NCO-4, respectively. (See also, Table A, below).

Table A

	Example 4	Comparative Experiment 1	Comparative Experiment 2
High molecular weight polyol (g) Polyol-1 Polyol-2	348.4	348.4	348.4
Carboxyl group-containing low molecular weight glycol (g) DMBA	25.8	25.8	25.8
Organic diisocyanate (g) IPDI	108.9	108.9	108.9
Carboxylic acid content of prepolymer (mmol/g)	0.35	0.35	0.35
Urethanes catalyst (g) DOTDL	0.04	0.04	0.04
Diluent (g) DMFDDG	224	224	224
Isocyanate content of prepolymer solution (%)	1.66	1.66	1.66
Nonionic polyisocyanate (g) NCO-2 NCO-3A NCO-4	100	100	100
Prepolymer/nonionic polyisocyanate	100/20	100/20	100/20
Neutralizing agent (g) TEA (added before emulsification)	17.6	17.6	17.6
Dispersion medium and chain extension agent (g) Water	1,000	1,000	1,000
Dispersion medium (g) Water	213	213	213
Amine aqueous solution (g) Chain extension agent IPDA Reaction terminating agent MEA	193.2 7.3	193.2 7.3	193.2 7.3
Production results Name of aqueous polyurethane emulsion Solid content (%) Viscosity at 25°C (mPa·s) Average particle diameter (nm)	PU-4 35.0 64 68	PU-10 35.5 649 133	PU-11 35.5 440 150

Production of Low-Functional Non-Yellowing Polyisocyanate

800 g of hexamethylene diisocyanate (HDI), 200 g of methoxypolyethylene glycol (MePEG-400) having a number average molecular weight of 400 and 0.1 g of zirconium octylate (manufactured by Daiichi Kigenso Kagaku Kogyo Co., Ltd.) were charged in a 1-L reactor vessel equipped with a stirrer, a thermometer, a nitrogen sealing tube and a condenser, and reaction was conducted at 110°C for 6 hours. Thereafter, 0.1 g of phosphoric acid was added thereto and termination reaction was conducted at 50°C for 1 hour. The isocyanate content of the reaction product after the termination reaction was 35.7%. This reaction product was then subjected to thin-film evaporation under conditions of 130°C and 0.04 kPa to obtain an allophanate-modified polyisocyanate (NCO-3A). The yield of NCO-3A was 37.1%. NCO-3A had an isocyanate content of 10.8 wt.%, a viscosity of 200 mPa·s at 25°C, a free diisocyanate content of 0.1 wt.%, and an average functional group number of 2. ¹H-NMR analysis of NCO-3A revealed that it had an isocyanurate content of 3 wt.%.

950 g of hexamethylene diisocyanate (HDI), 50 g of isopropyl alcohol and 0.1 g of zirconium octylate (manufactured by Daiichi Kigenso Kagaku Kogyo Co., Ltd.) were charged in a 1-L reactor vessel equipped with a stirrer, a thermometer, a nitrogen sealing tube and a condenser, and reaction was conducted at 110°C for 6 hours. Thereafter, 0.1 g of phosphoric acid was added thereto and termination reaction was conducted at 50°C for 1 hour. The isocyanate content of the reaction product after the termination reaction was 42.1%. This reaction product was then subjected to thin-film evaporation under conditions of 130°C and 0.04 kPa to obtain an allophanate-modified polyisocyanate (NCO-3B). The yield of NCO-3B was

30%. NCO-3B had an isocyanate content of 21.1 wt.%, a viscosity of 118 mPa·s at 25°C, a free diisocyanate content of 0.1 wt.%, and an average functional group number of 2. ¹H-NMR analysis of NCO-3B revealed that it had an isocyanurate content of 3 wt.%.

280 g of NCO-3A and 780 g of NCO-3B were mixed to obtain NCO-4.

Evaluation of Coated Film

Next, coated film evaluations were carried out with regard to each of the obtained PU-4 (reproduced), PU-10 and PU-11. The evaluations were carried out in accordance with the methods disclosed in paragraph [0068] of the specification. The results thereof are shown in Table B, below.

Additionally, evaluations were also made under other conditions. Specifically, films obtained through drying at 150°C were subjected to evaluations of solvent resistance and alkali resistance. The alkali resistance evaluation was made in accordance with the manner as disclosed in the specification (page 33) except for replacing the 5% sodium carbonate aqueous solution with a 5% sodium hydroxide aqueous solution. The solvent resistance evaluation was made in the same manner as disclosed in the specification (page 33) or using 50% ethanol in place of MEK. The numbers shown in connection with the solvent resistance denote the number of rubbing at the time when occurrence of change such as whitening was observed.

Table B

	Example 4'	Comparative Experiment 1	Comparative Experiment 2
Polyisocyanate component			
NCO%	16.8	10.5	19.5
Average functional group number	3.2	2	2
EO%	13.6	49.6	13.6
Dispersion			
Particle diameter (nm)	68	133	160
Viscosity (mPa.s at 25°C)	64	649	440
Solid content	35.5	35.5	35.5
Evaluation results of coated film			
(Film dried at 160°C)			
Solvent resistance (50% ethanol)	30	10	15
Solvent resistance (MEK)	30	8	16
Alkali resistance	A	B	B
(Film dried at 220°C)			
Appearance	A	A-B	B
Solvent resistance (MEK)	A (>200)	B (80)	B (120)

Conclusion

Comparing the coated film of the polyurethane emulsion using a trifunctional isocyanate with the coated films of the polyurethane emulsions using a bifunctional isocyanate, the former is superior in solvent (MEK) resistance and alkali resistance, as compared to the latter. Further, the polyurethane emulsions using a bifunctional isocyanate resulted in an increased particle size, which led to a deteriorated appearance of the obtained coated film.

Therefore, unexpected substantially improved results were obtained in an embodiment of the present invention in accordance with Example 4 of the present specification wherein the trifunctional nonionic isocyanate was replaced with a bifunctional isocyanate.

I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are

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U.S. Application No.: 10/591,422

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punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: 9/26/07

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